Absorption by ambient aerosols during CalNex

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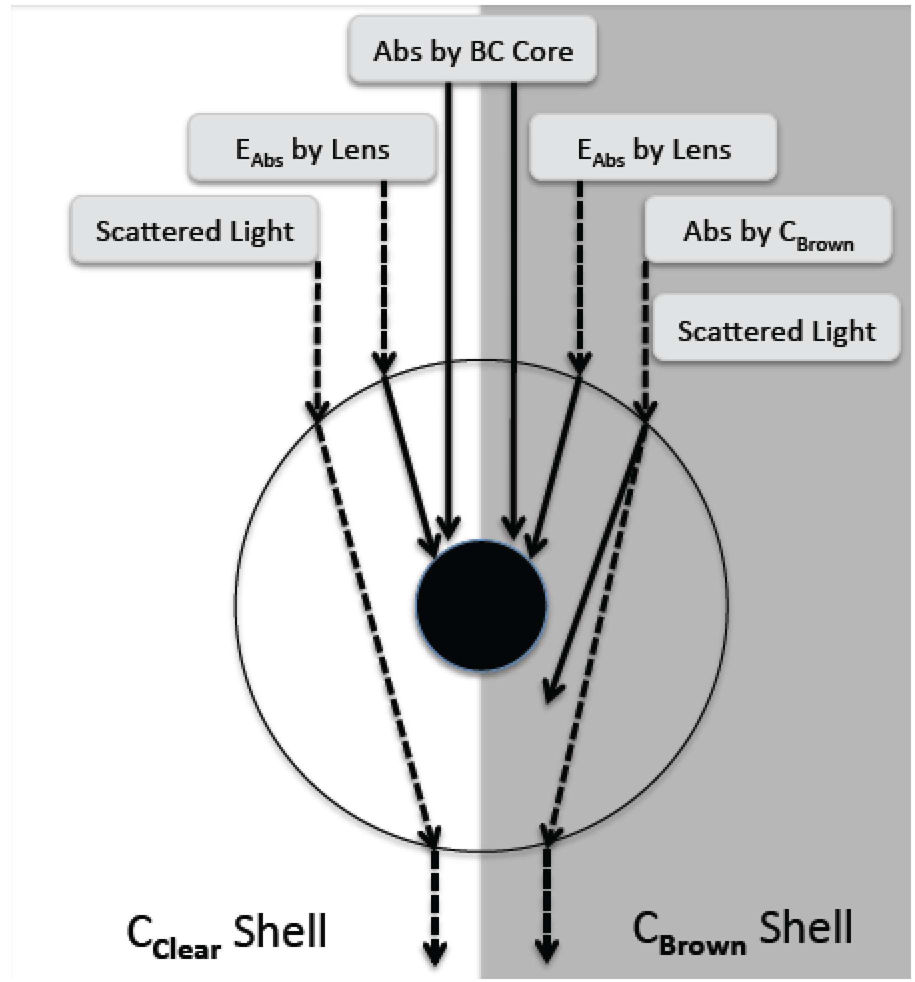
Aerosol Volatility and Optical Properties

Light absorption by particles from **black** carbon (soot) and **brown** carbon

Coatings on BC can theoretically ***enhance*** absorption in climatically important ways
[e.g. Jacobson, *Nature*, 2001]

Enhancement factor can be calculated from Mie theory, but limited validation from *ambient* measurements

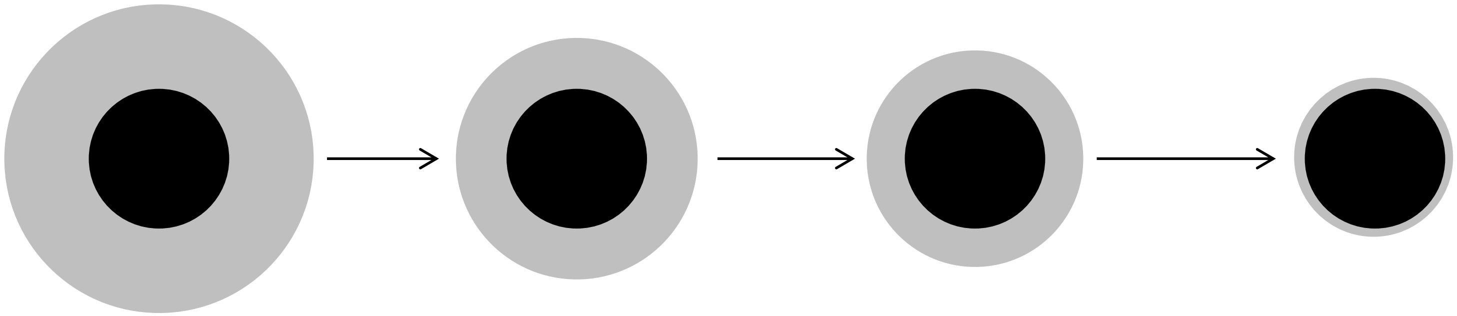
Objective: to directly measure the absorption enhancement factor for ambient particles at multiple wavelengths



[Lack and Cappa, *ACP*, 2010]

Approach: Heat Particles to Drive Off Coatings

Ambient → Really hot

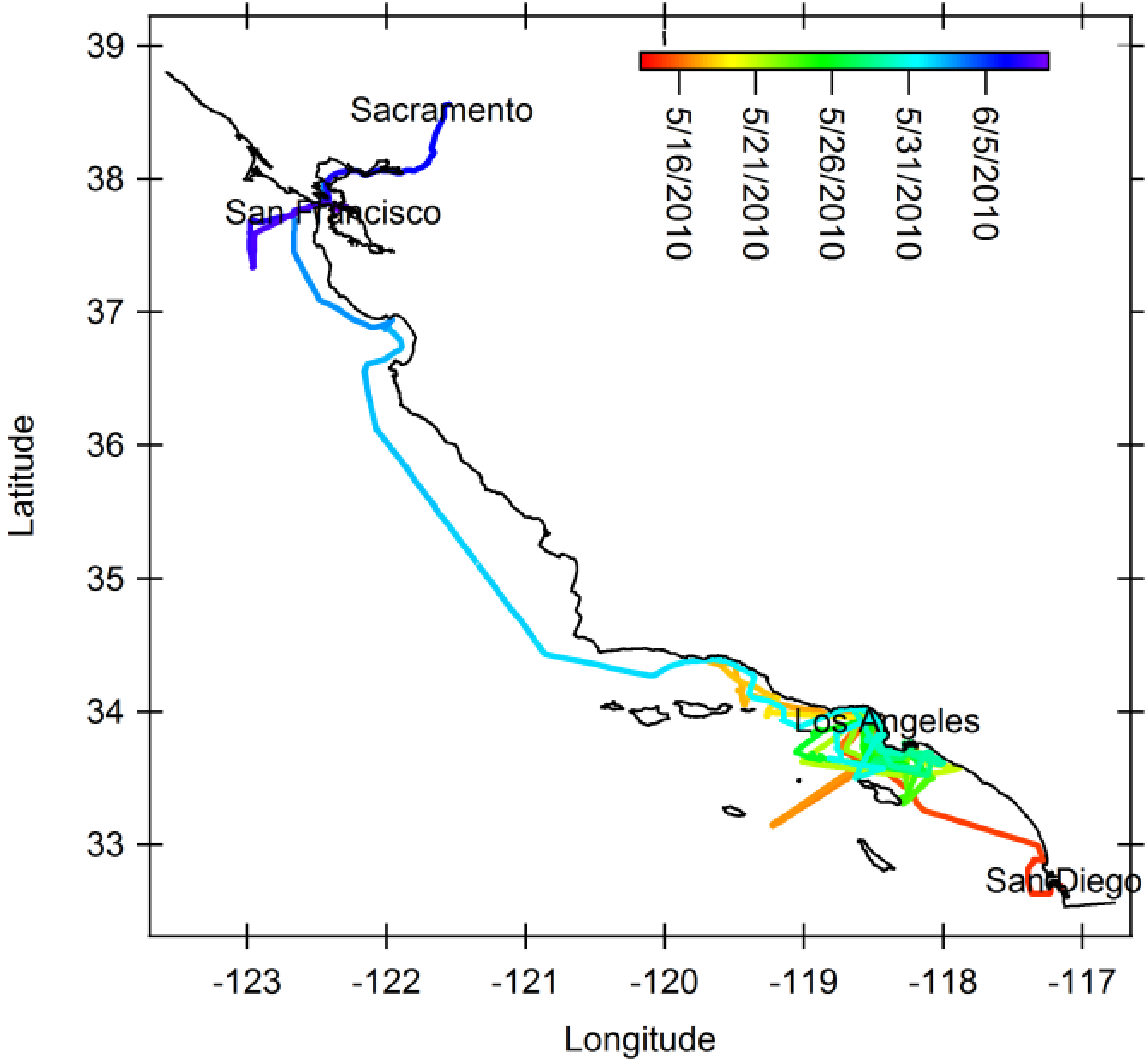


Large Enhancement → Small Enhancement

Highly Scattering → Less Scattering

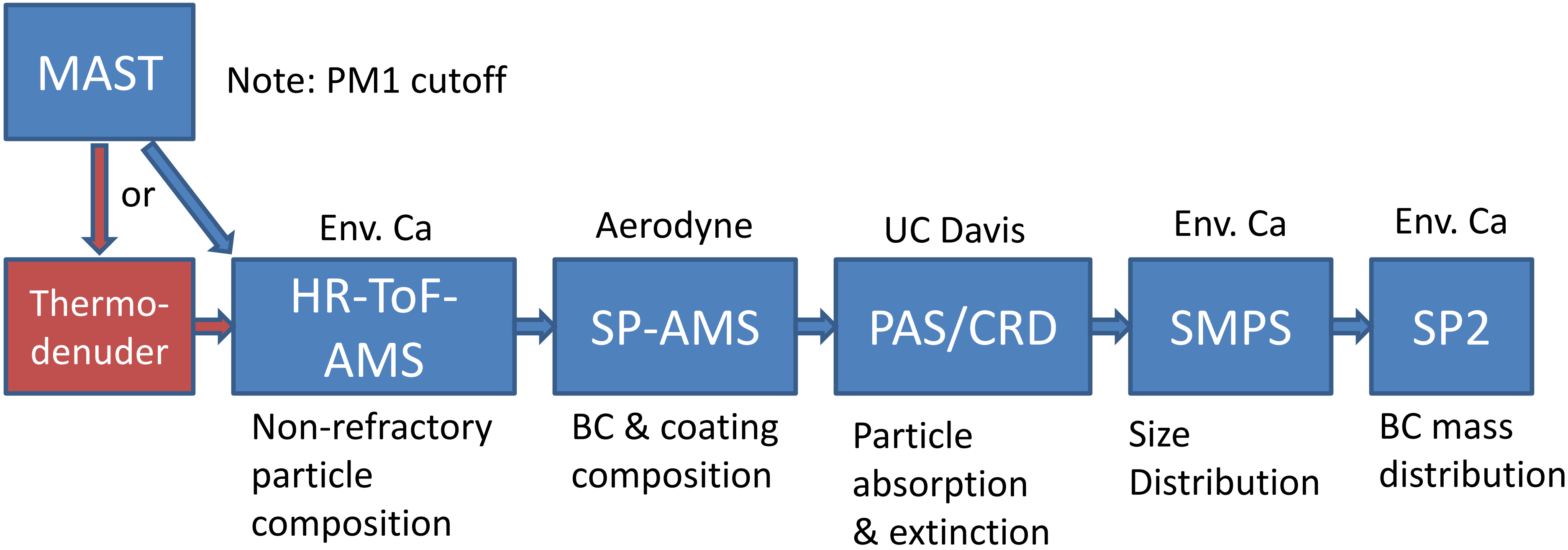
Brown Carbon → No Brown Carbon

Absorption by brown carbon should contribute more to total absorption at short λ



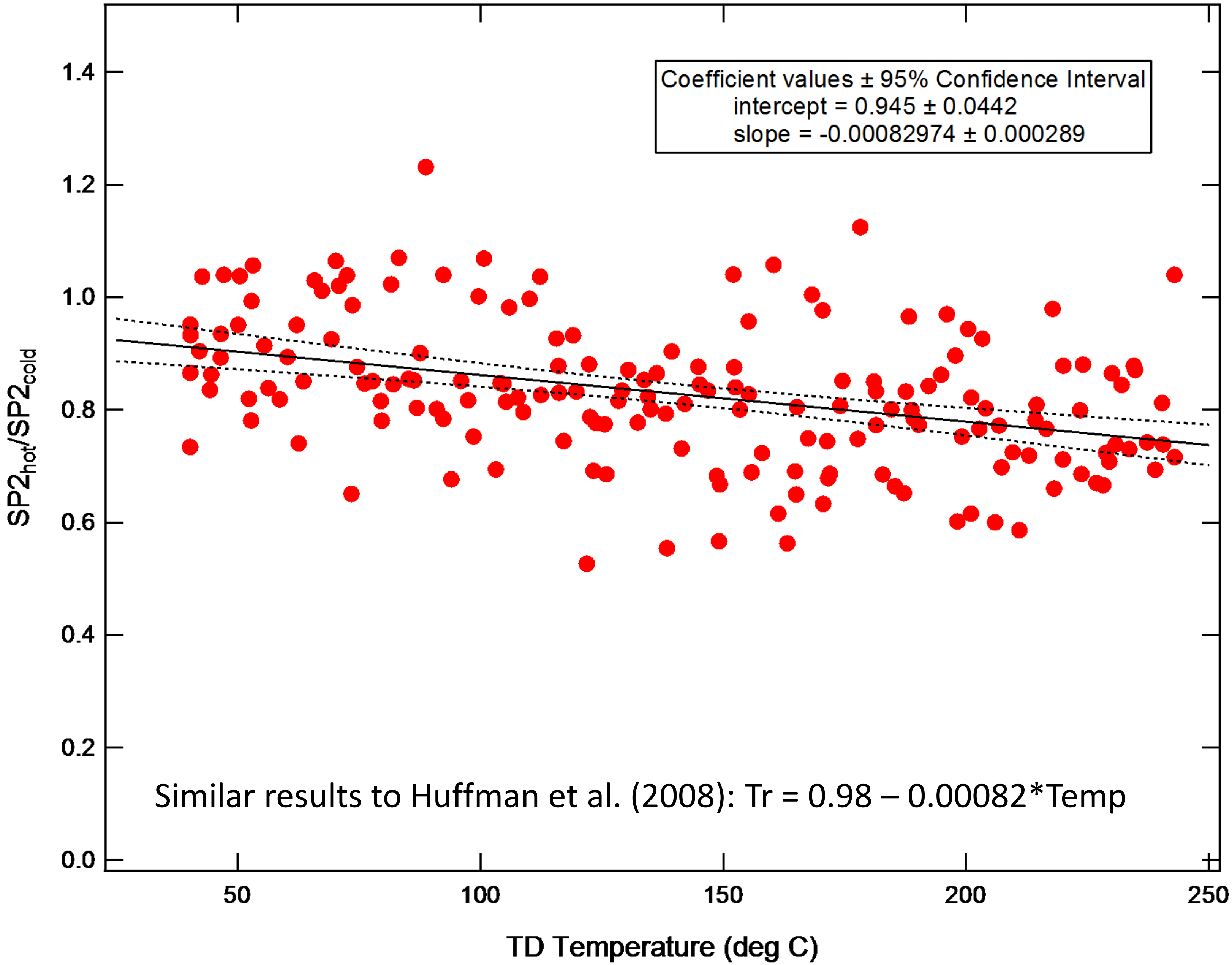
Sacramento
San Francisco
Los Angeles
San Diego
5/16/2010
5/21/2010
5/26/2010
5/31/2010
6/5/2010
39
38
37
36
35
34
33
Latitude
-123
-122
-121
-120
-119
-118
-117
Longitude

Measurements

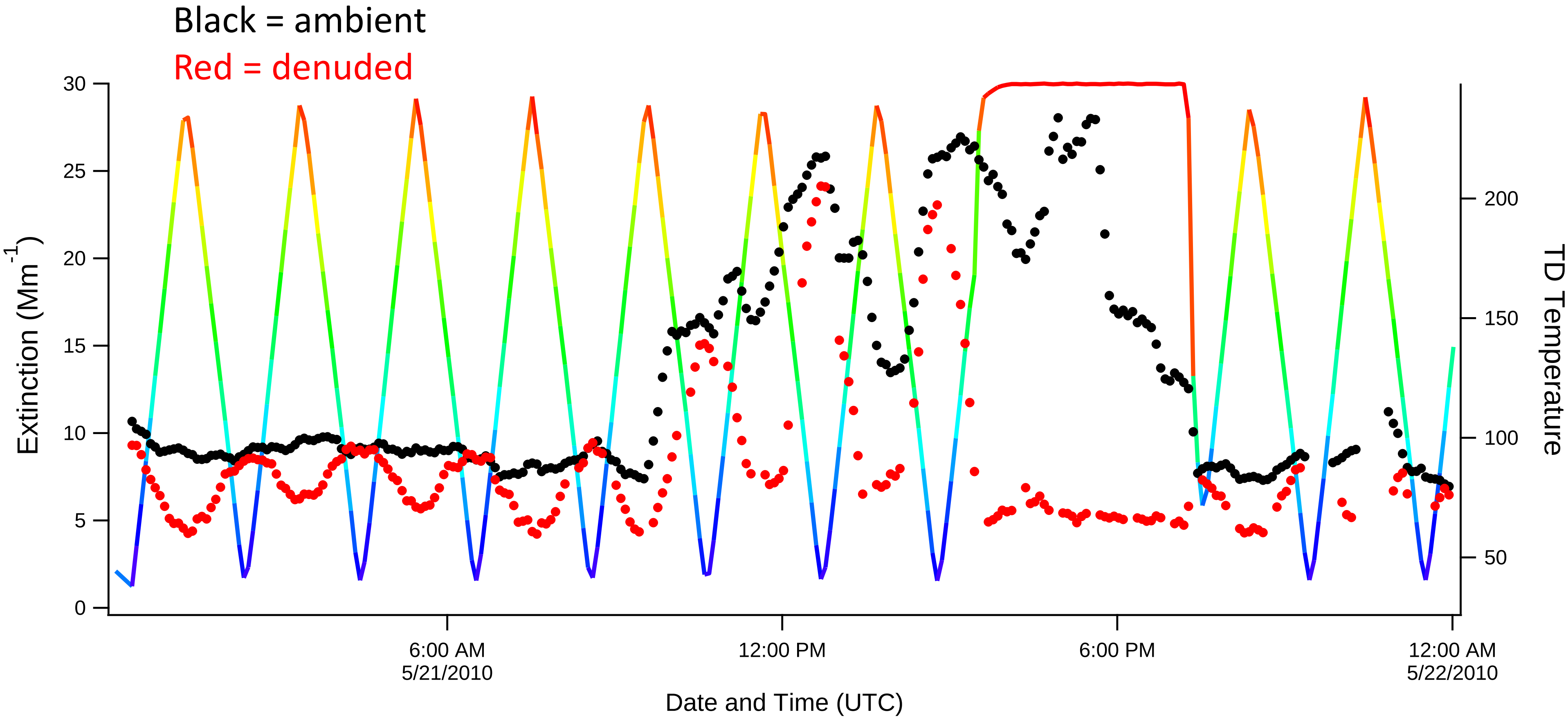


- **PAS/CRD** measures light absorption and light extinction at 532 nm and 405 nm
- **SP-AMS** measures BC mass and coating composition
- **SP2** measures BC mass and infers coating thickness
- **SP2** and **SPAMS** tell us something about how coating thickness has changed upon heating
- **SP2** BC measurement can tell us about particle losses through the TD

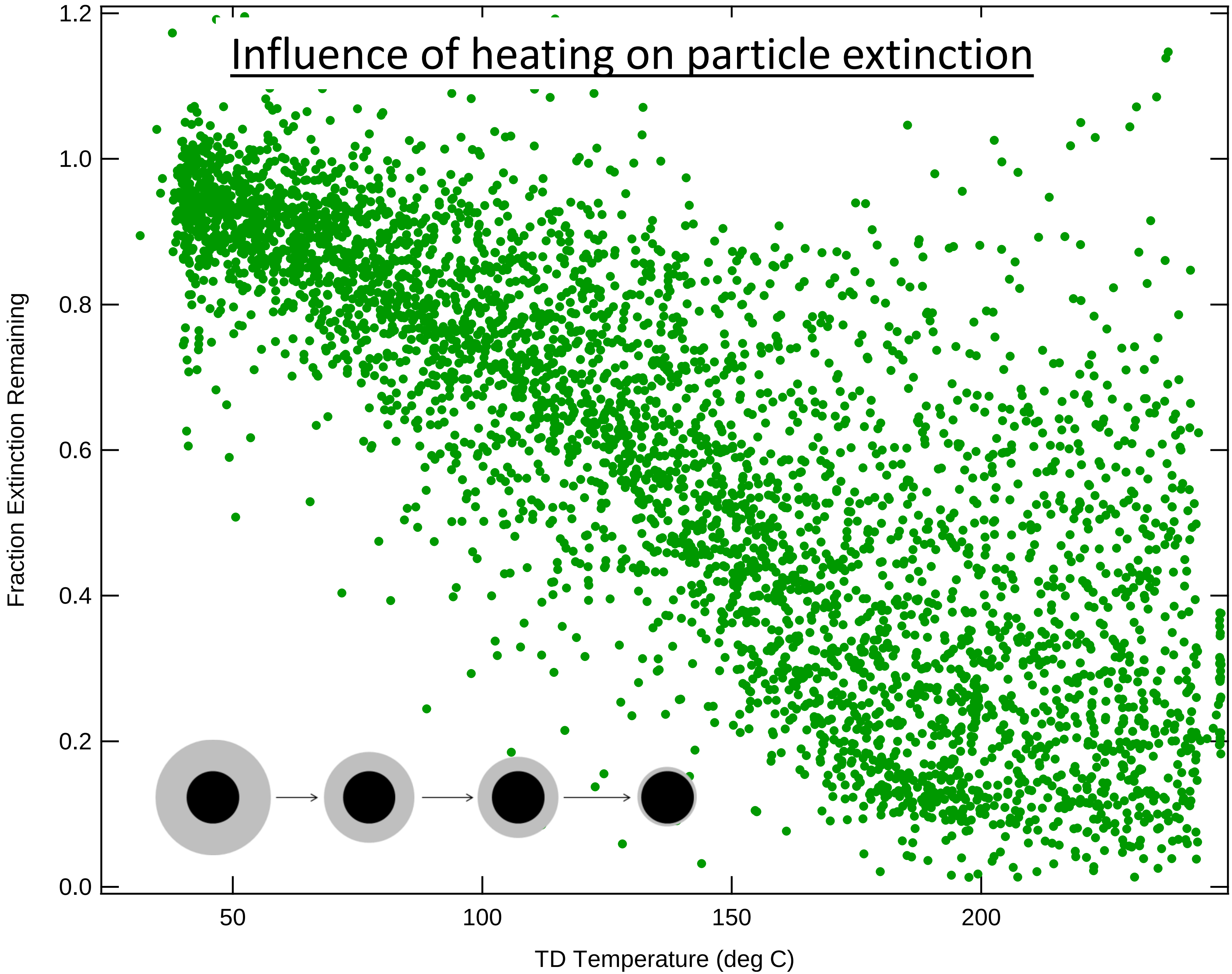
Particle Transmission Correction

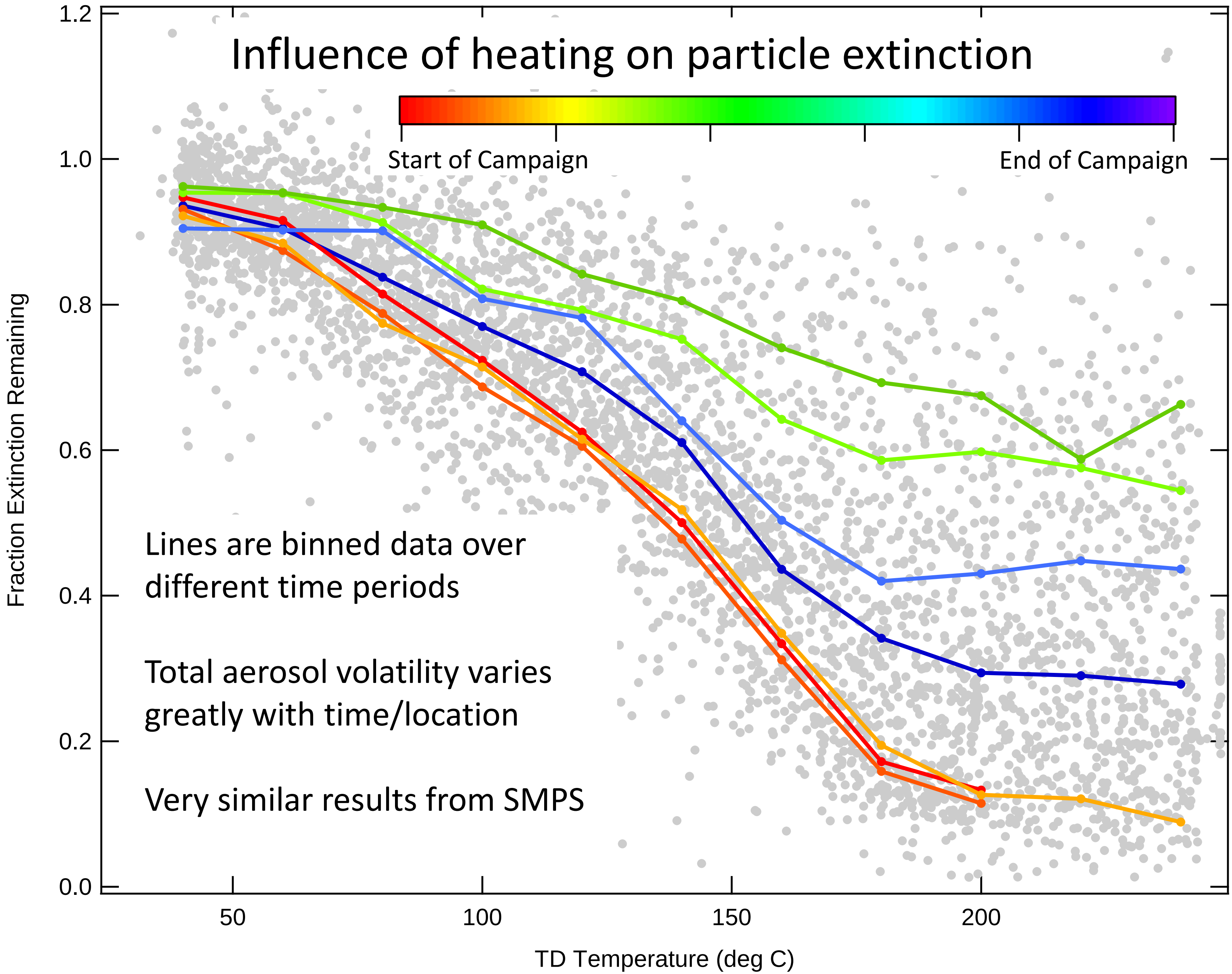


Influence of heating on particle extinction (532 nm)



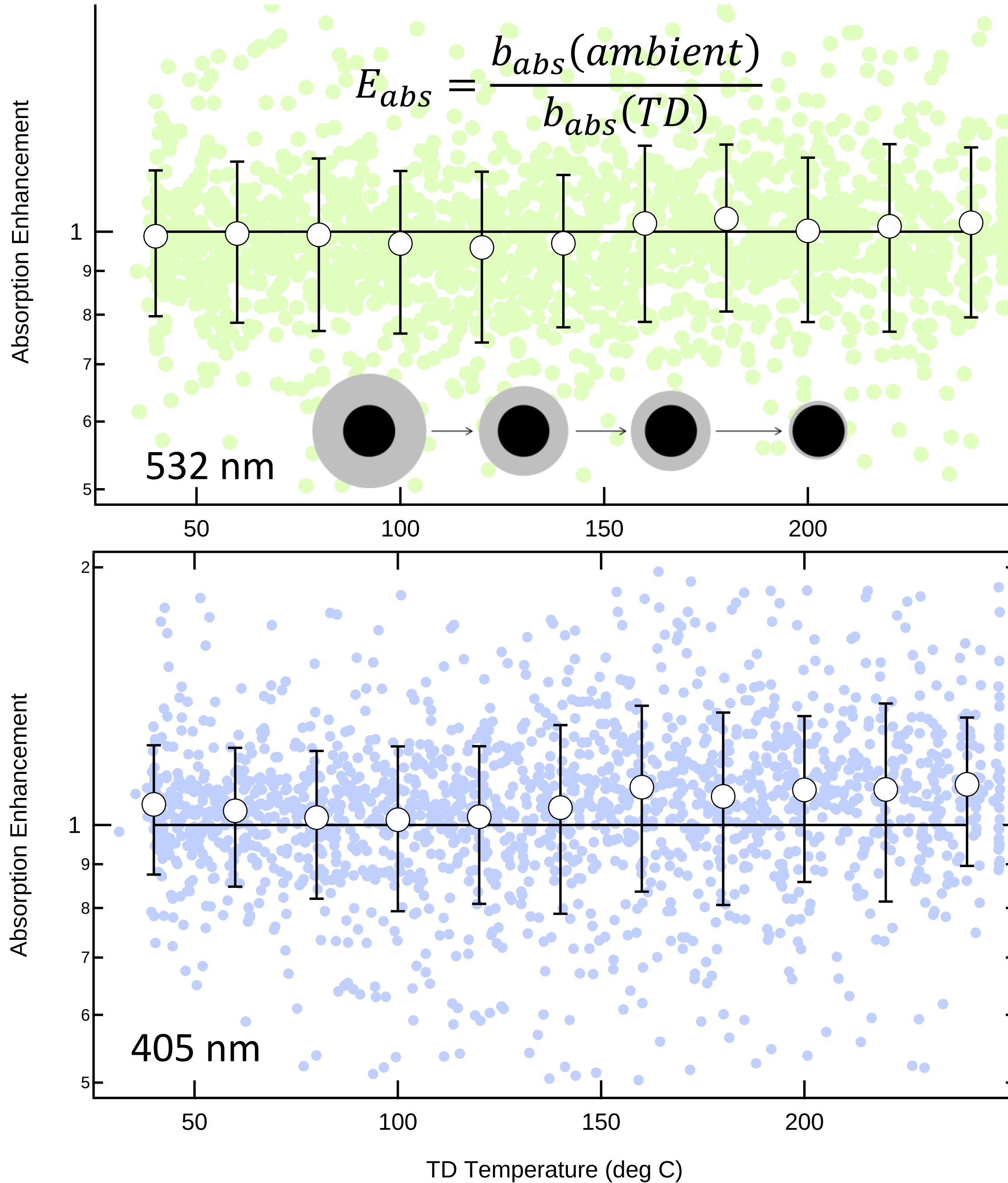
$$Extinction\ Fraction\ Remaining = \frac{\sigma_{ext}(T)}{\sigma_{ext}(ambient)}$$


Influence of heating on particle extinction
Fraction Extinction Remaining
TD Temperature (deg C)
1.2
1.0
0.8
0.6
0.4
0.2
0.0
50
100
150
200



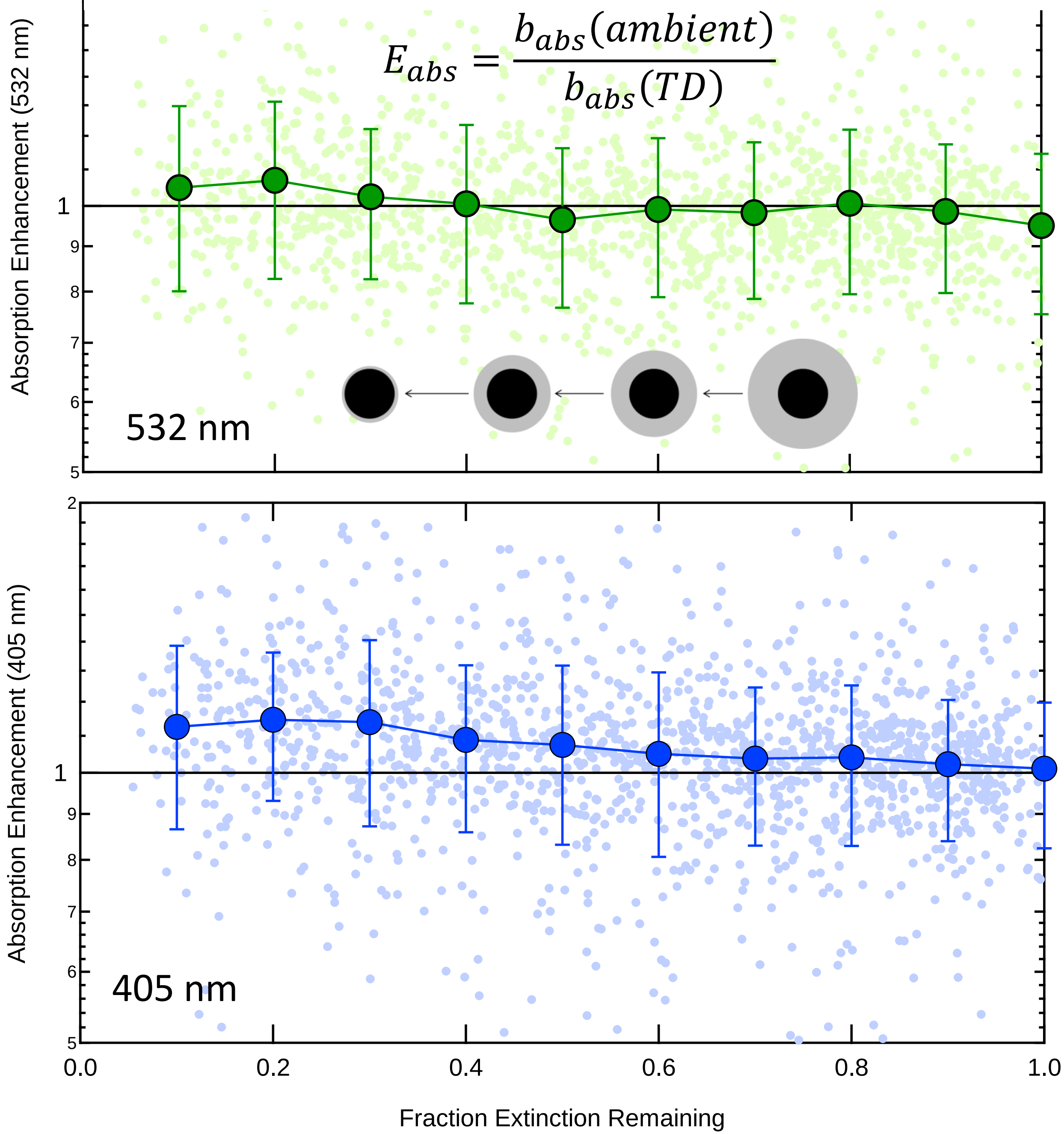
Influence of heating on particle extinction
Start of Campaign
End of Campaign
Lines are binned data over different time periods
Total aerosol volatility varies greatly with time/location
Very similar results from SMPS
Fraction Extinction Remaining
TD Temperature (deg C)
0.0
0.2
0.4
0.6
0.8
1.0
1.2
50
100
150
200

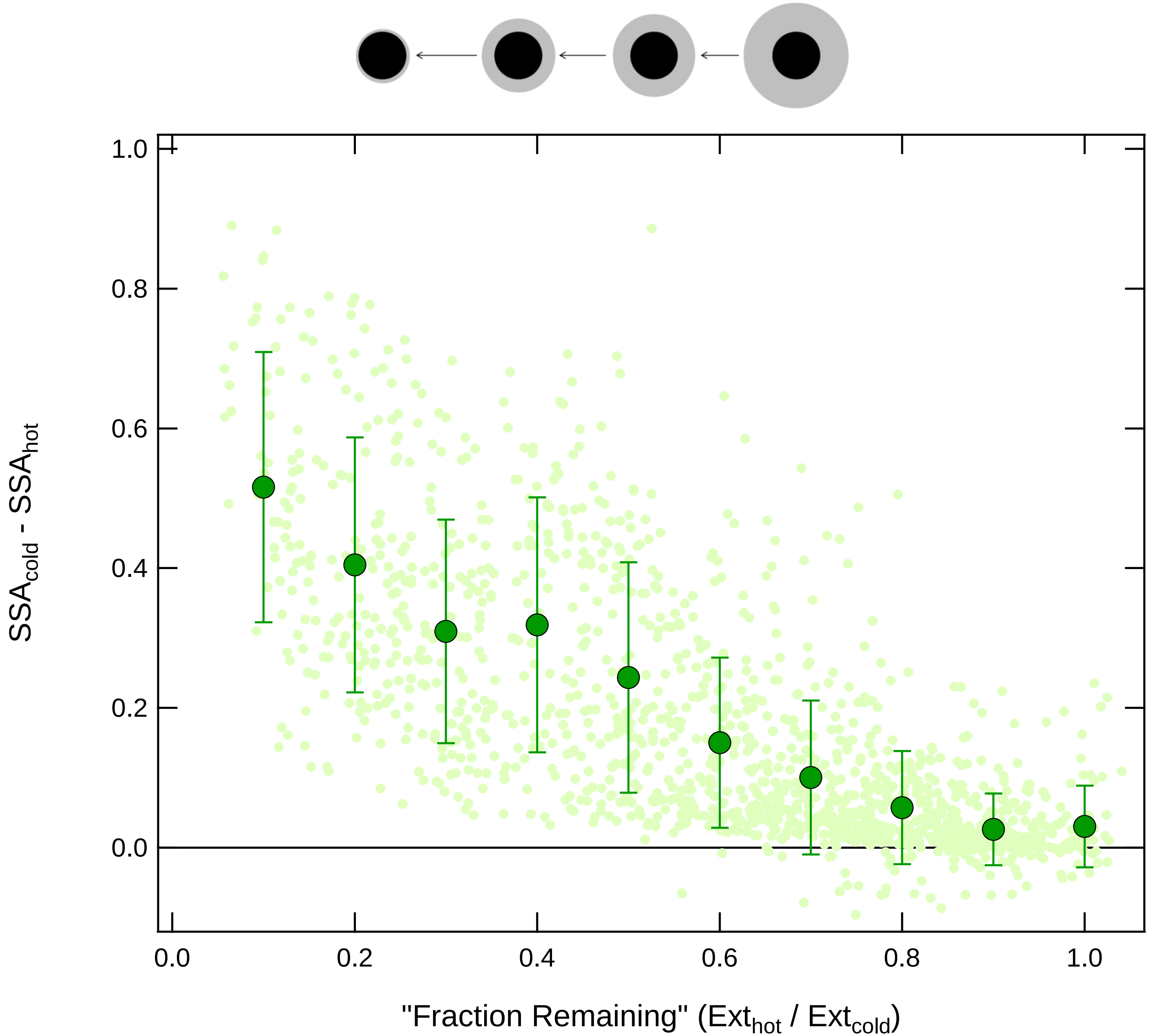
Influence of heating on particle absorption



Note: only include periods when $b_{abs,ambient} > 0.5$ Mm^{-1}

Influence of heating on particle absorption

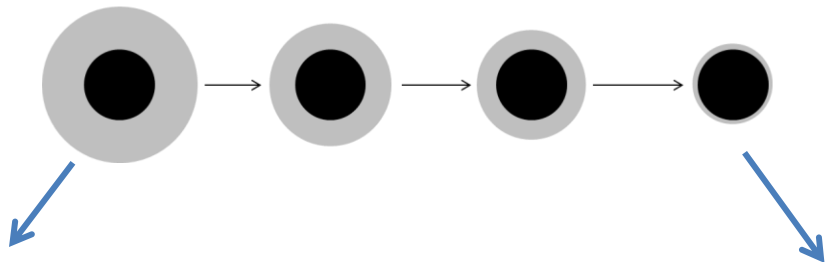




1.0
0.8
0.6
0.4
0.2
0.0
SSA_cold - SSA_hot
0.0
0.2
0.4
0.6
0.8
1.0
"Fraction Remaining" (Ext_hot / Ext_cold)

Theoretical Calculations of Aerosol Optical Properties

Use "core-shell" Mie theory to calculate evolution of σ_{abs}, σ_{ext} and SSA



D_p(particle) = 400 nm
D_p(core) = 80 nm
$\sigma_{abs} = 9.8 \times 10^{-15}$ m^2/particle
$\sigma_{ext} = 2.9 \times 10^{-13}$ m^2/particle
SSA = 0.97

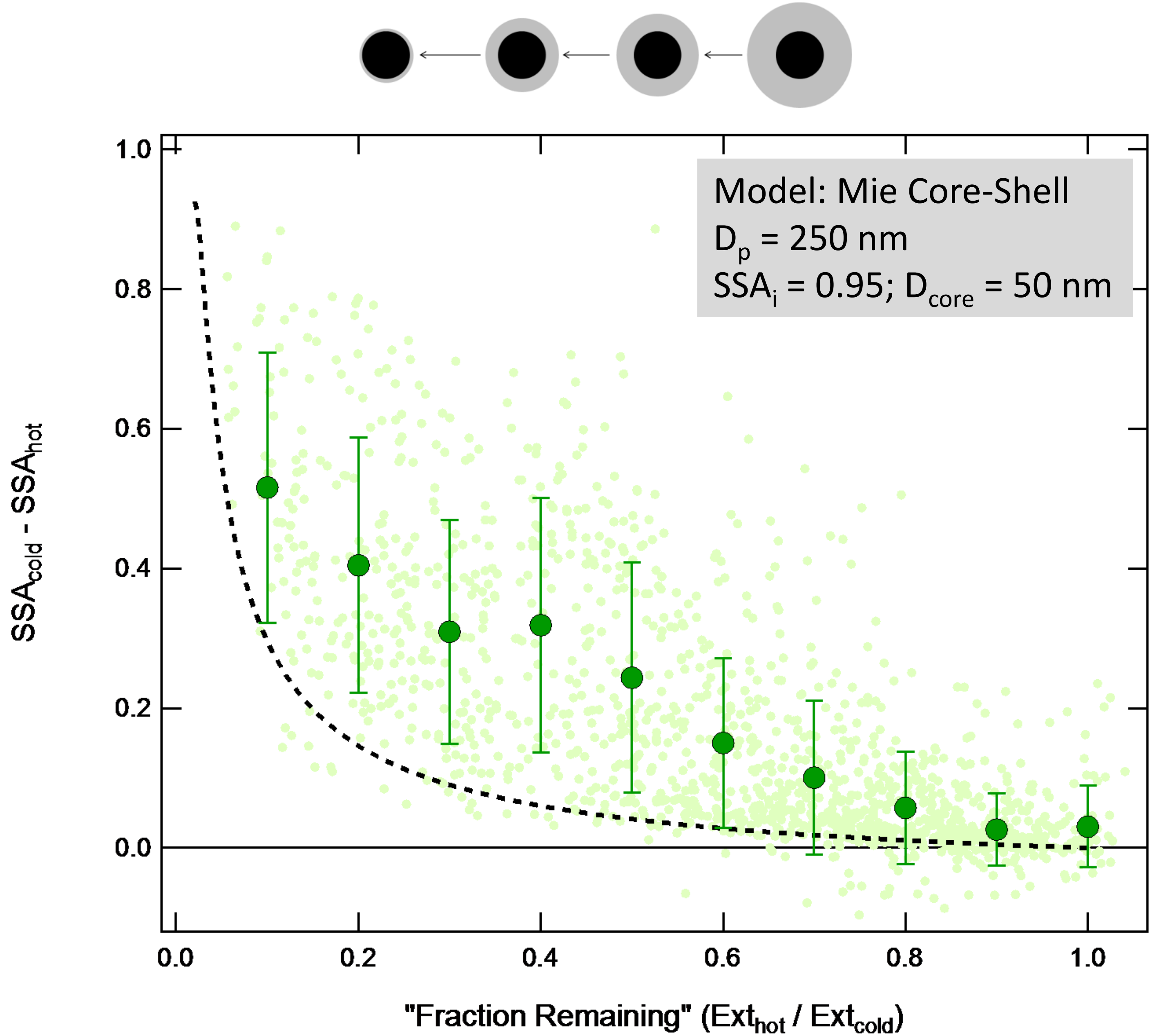
E_{abs} = 2.57
FR_{ext} = 0.015
ΔSSA = 0.84

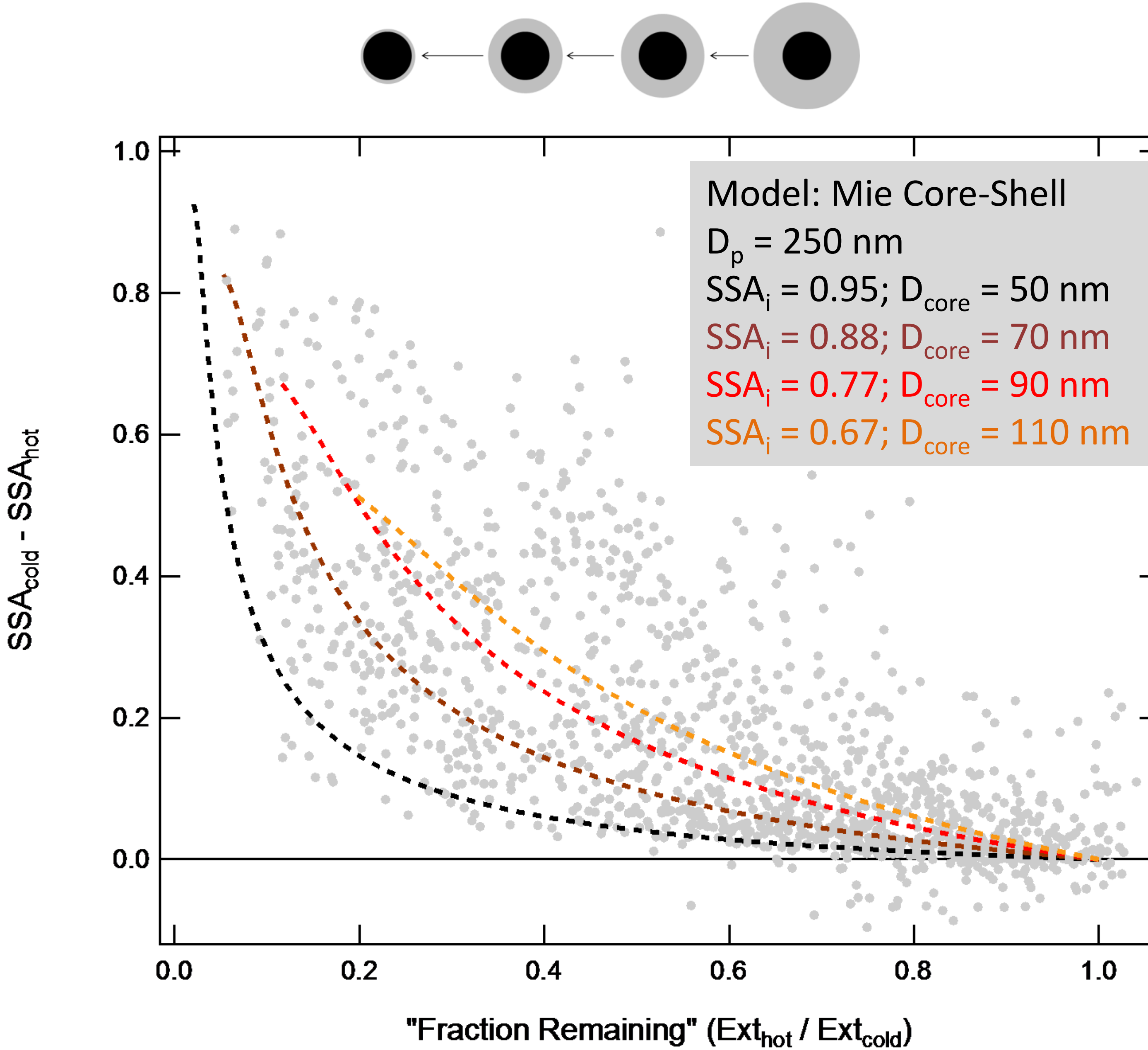
D_p(particle) = 100 nm
D_p(core) = 80 nm
$\sigma_{abs} = 3.8 \times 10^{-15}$ m^2/particle
$\sigma_{ext} = 4.4 \times 10^{-15}$ m^2/particle
SSA = 0.13

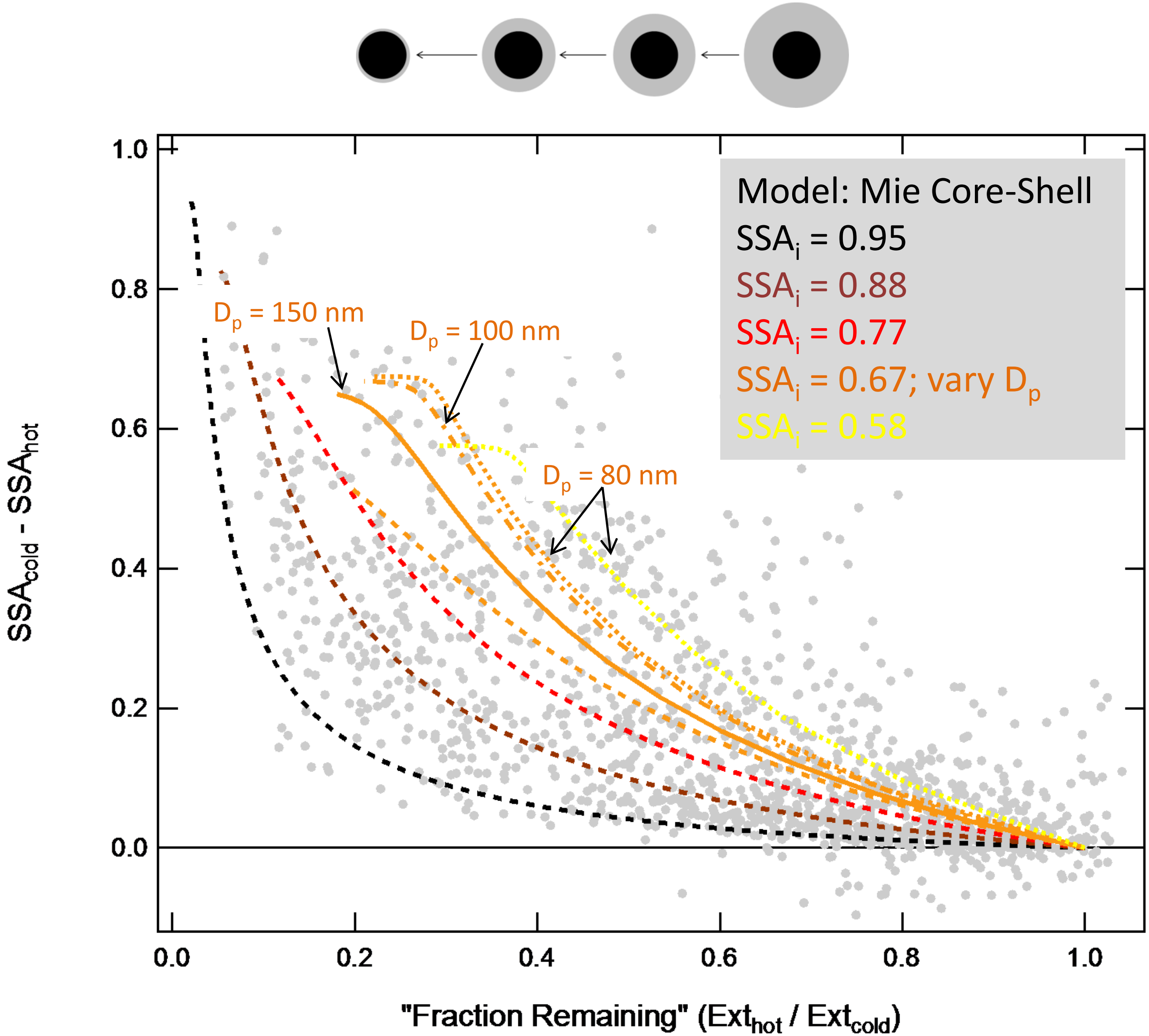
D_p(particle) = 200 nm
D_p(core) = 60 nm
$\sigma_{abs} = 2.9 \times 10^{-15}$ m^2/particle
$\sigma_{ext} = 1.5 \times 10^{-14}$ m^2/particle
SSA = 0.81

E_{abs} = 1.45
FR_{ext} = 0.19
ΔSSA = 0.65

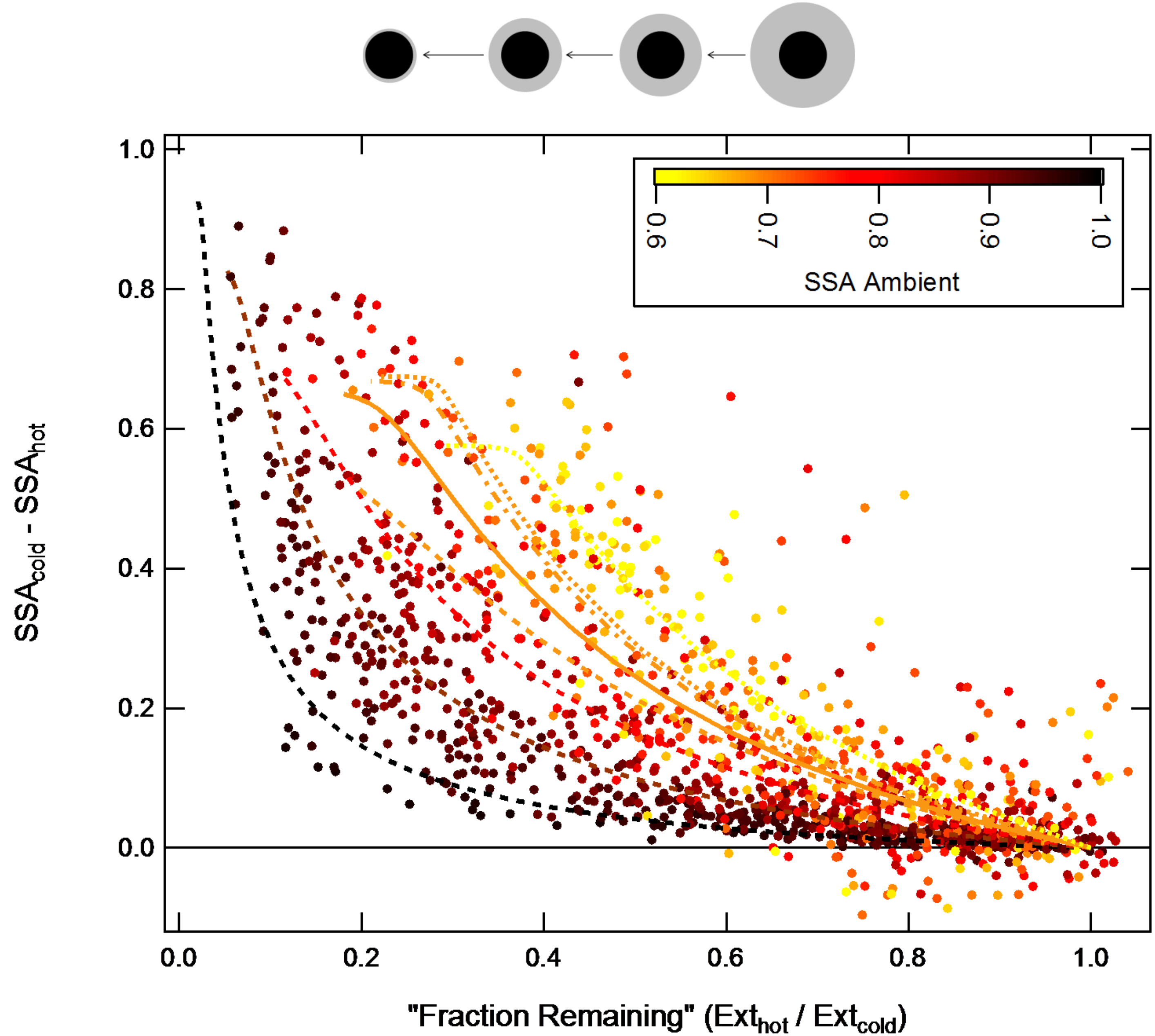
D_p(particle) = 120 nm
D_p(core) = 60 nm
$\sigma_{abs} = 2.0 \times 10^{-15}$ m^2/particle
$\sigma_{ext} = 2.8 \times 10^{-15}$ m^2/particle
SSA = 0.16


Model: Mie Core-Shell
D_p = 250 nm
SSA_i = 0.95; D_{core} = 50 nm
SSA_{cold} - SSA_{hot}
"Fraction Remaining" (Ext_{hot} / Ext_{cold})
1.0
0.8
0.6
0.4
0.2
0.0
0.0
0.2
0.4
0.6
0.8
1.0

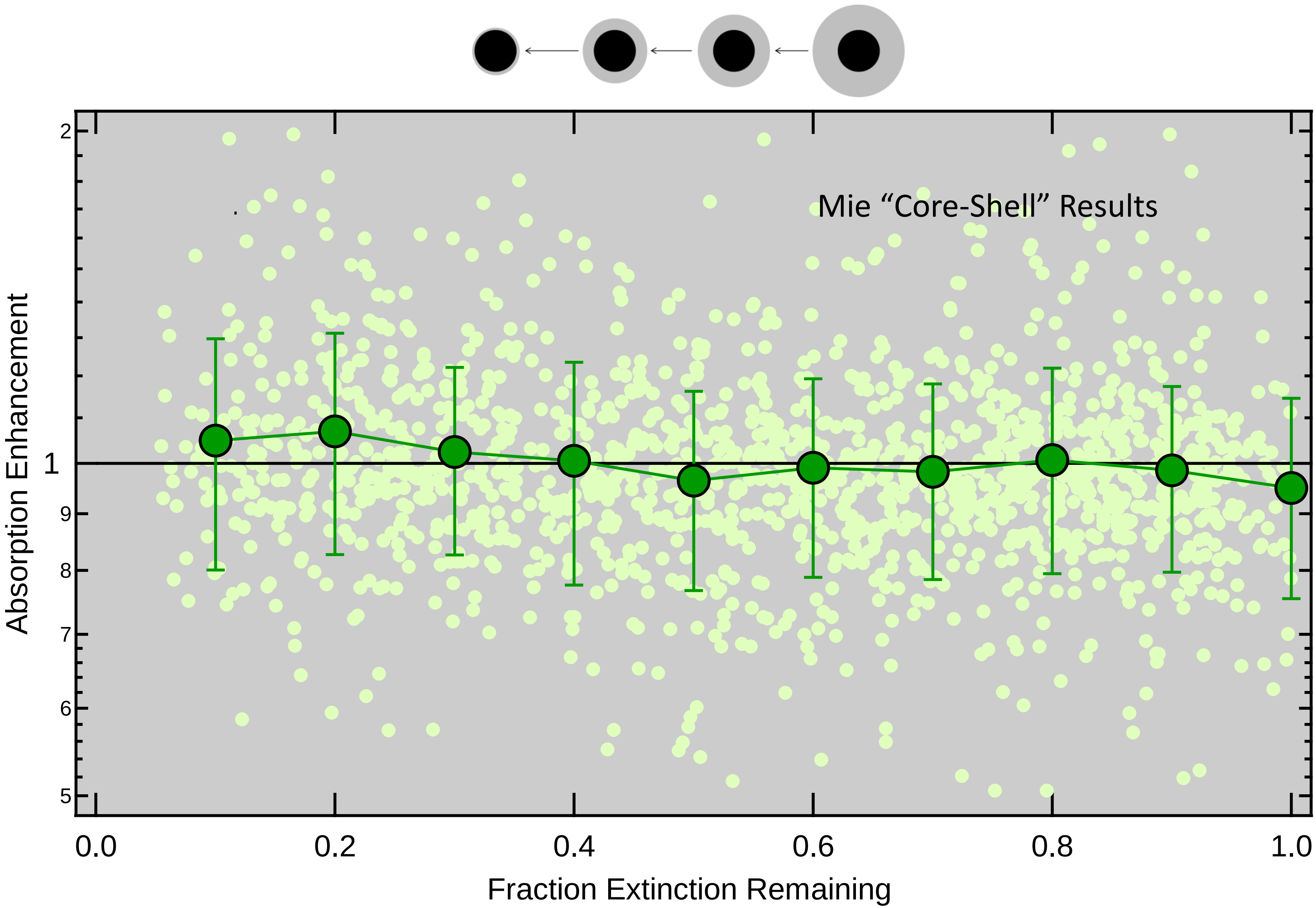

Model: Mie Core-Shell
D_p = 250 nm
SSA_i = 0.95; D_{core} = 50 nm
SSA_i = 0.88; D_{core} = 70 nm
SSA_i = 0.77; D_{core} = 90 nm
SSA_i = 0.67; D_{core} = 110 nm
1.0
0.8
0.6
0.4
0.2
0.0
SSA_{cold} - SSA_{hot}
0.0
0.2
0.4
0.6
0.8
1.0
"Fraction Remaining" (Ext_{hot} / Ext_{cold})



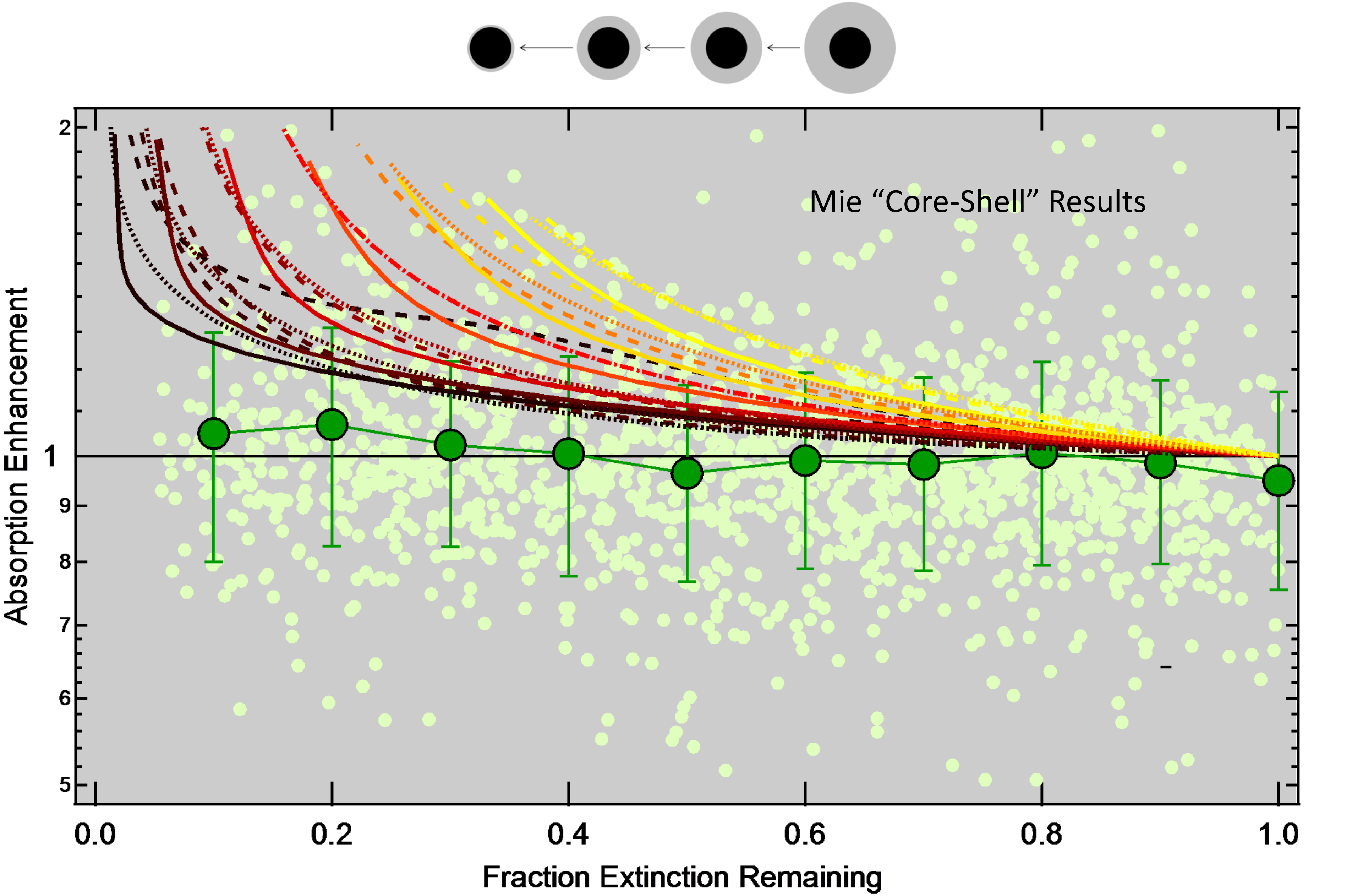
Model: Mie Core-Shell
SSA_i = 0.95
SSA_i = 0.88
SSA_i = 0.77
SSA_i = 0.67; vary D_p
SSA_i = 0.58
D_p = 150 nm
D_p = 100 nm
D_p = 80 nm
SSA_{cold} - SSA_{hot}
"Fraction Remaining" (Ext_{hot} / Ext_{cold})
0.0
0.2
0.4
0.6
0.8
1.0



SSA Ambient
0.6
0.7
0.8
0.9
1.0
SSAcold - SSAhot
"Fraction Remaining" (Exthot / Extcold)
0.0
0.2
0.4
0.6
0.8
1.0



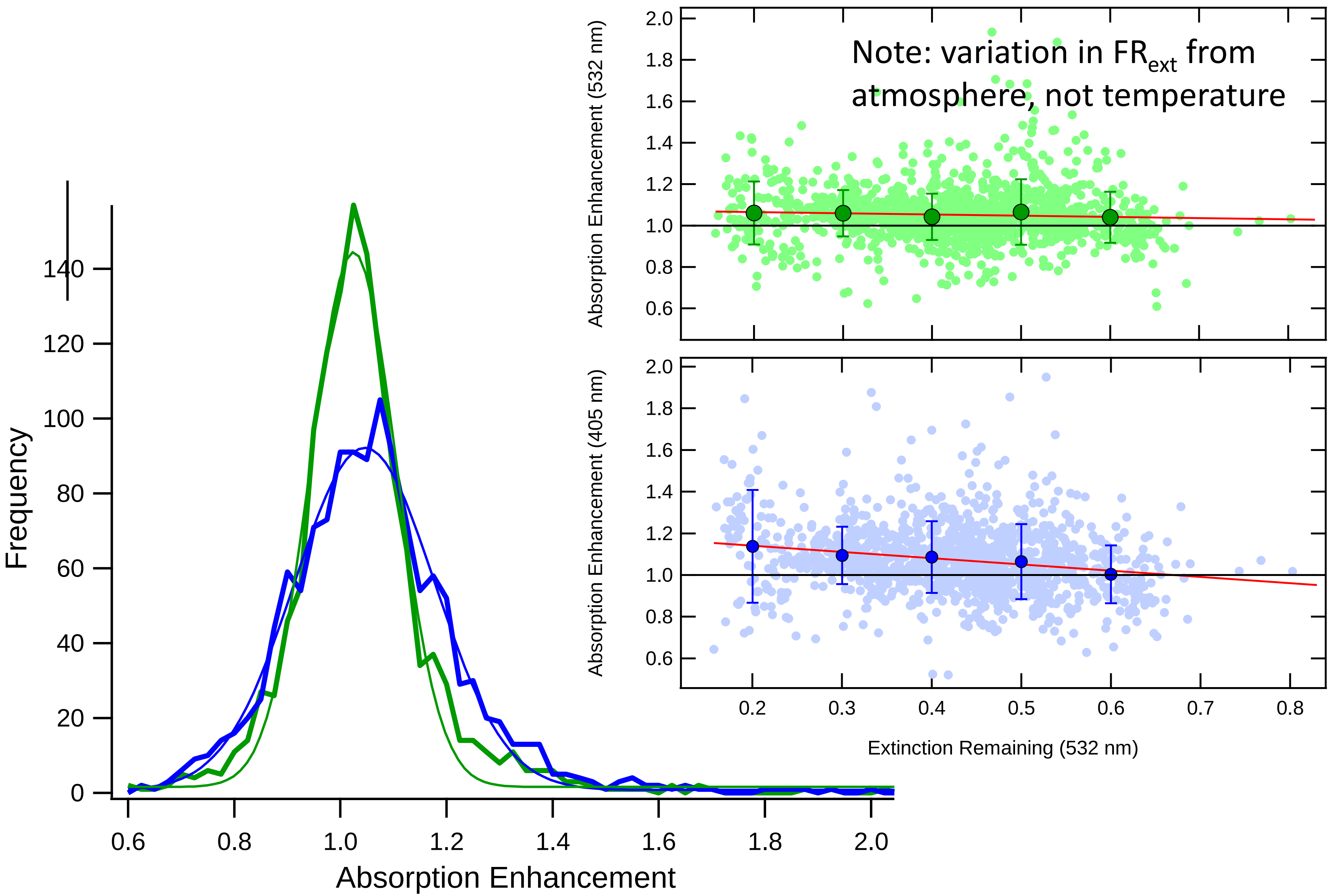
Mie "Core-Shell" Results
Absorption Enhancement
2
1
9
8
7
6
5
0.0
0.2
0.4
0.6
0.8
1.0
Fraction Extinction Remaining



Mie "Core-Shell" Results
Absorption Enhancement
2
1
9
8
7
6
5
0.0
0.2
0.4
0.6
0.8
1.0
Fraction Extinction Remaining

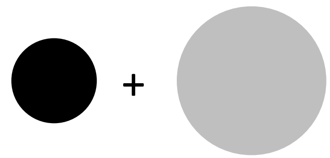
Since we are talking about California anyway…

same experiment done at CARES, but with fixed TD temperature (225 °C) and $PM_{2.5}$

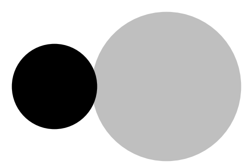


Why such an (unexpectedly) low absorption enhancement?

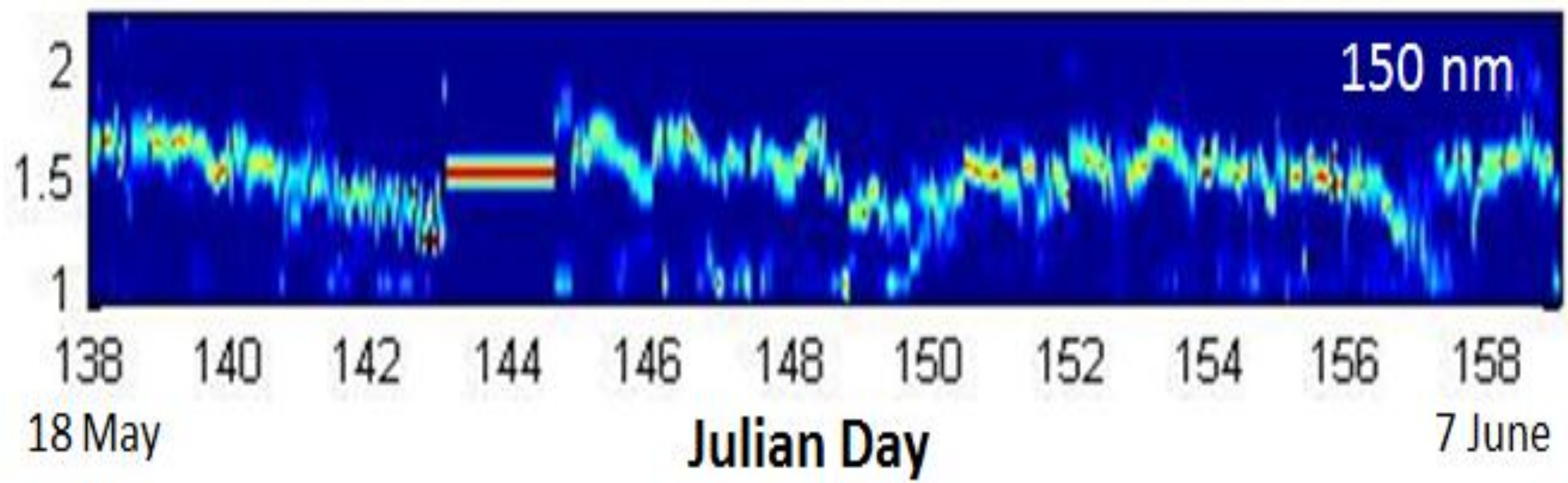
1. Concern: particles are not internally mixed

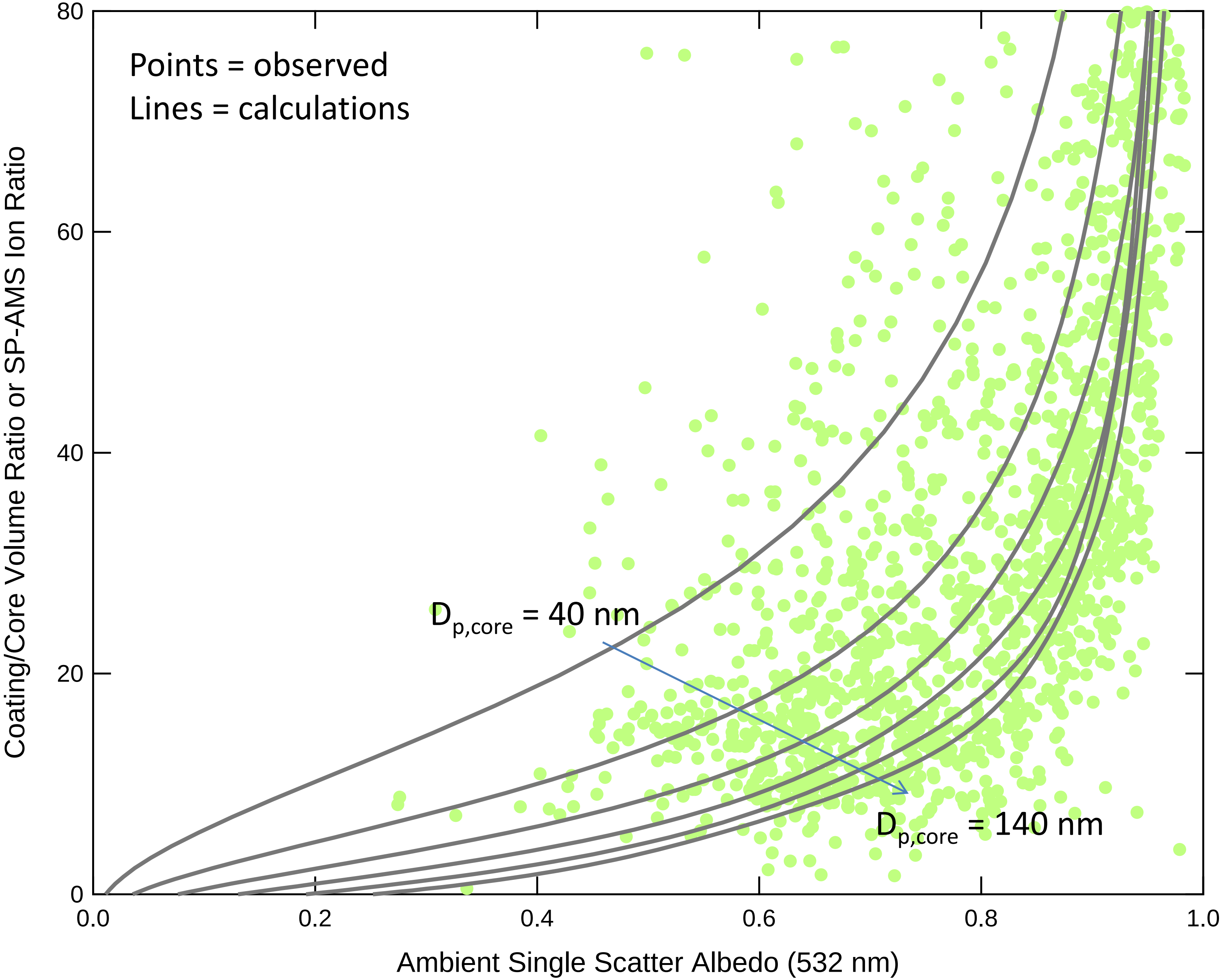


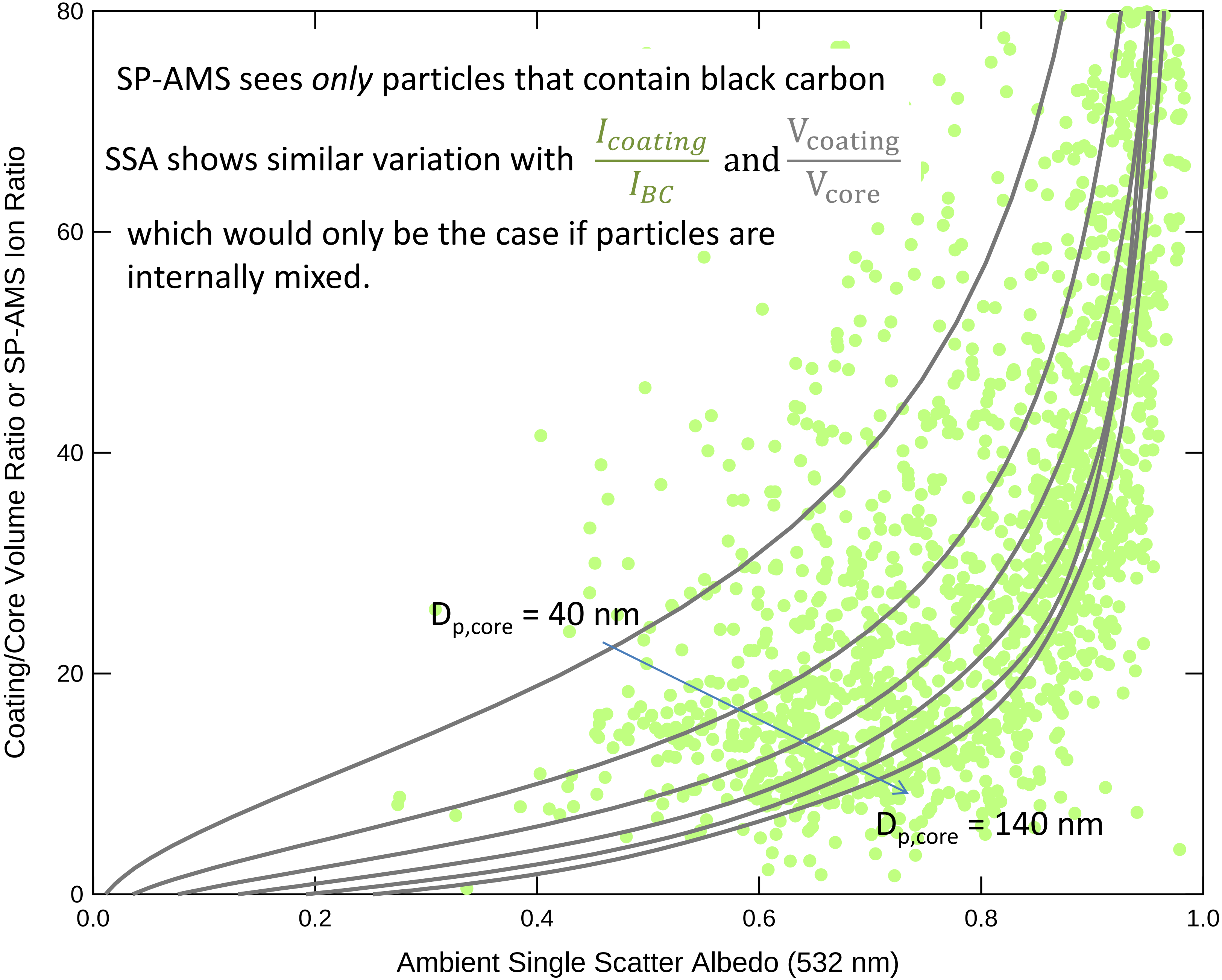
2. Particles are internally mixed, but not with “core-shell” morphology



H-TDMA indicates aerosol dominated by single growth mode

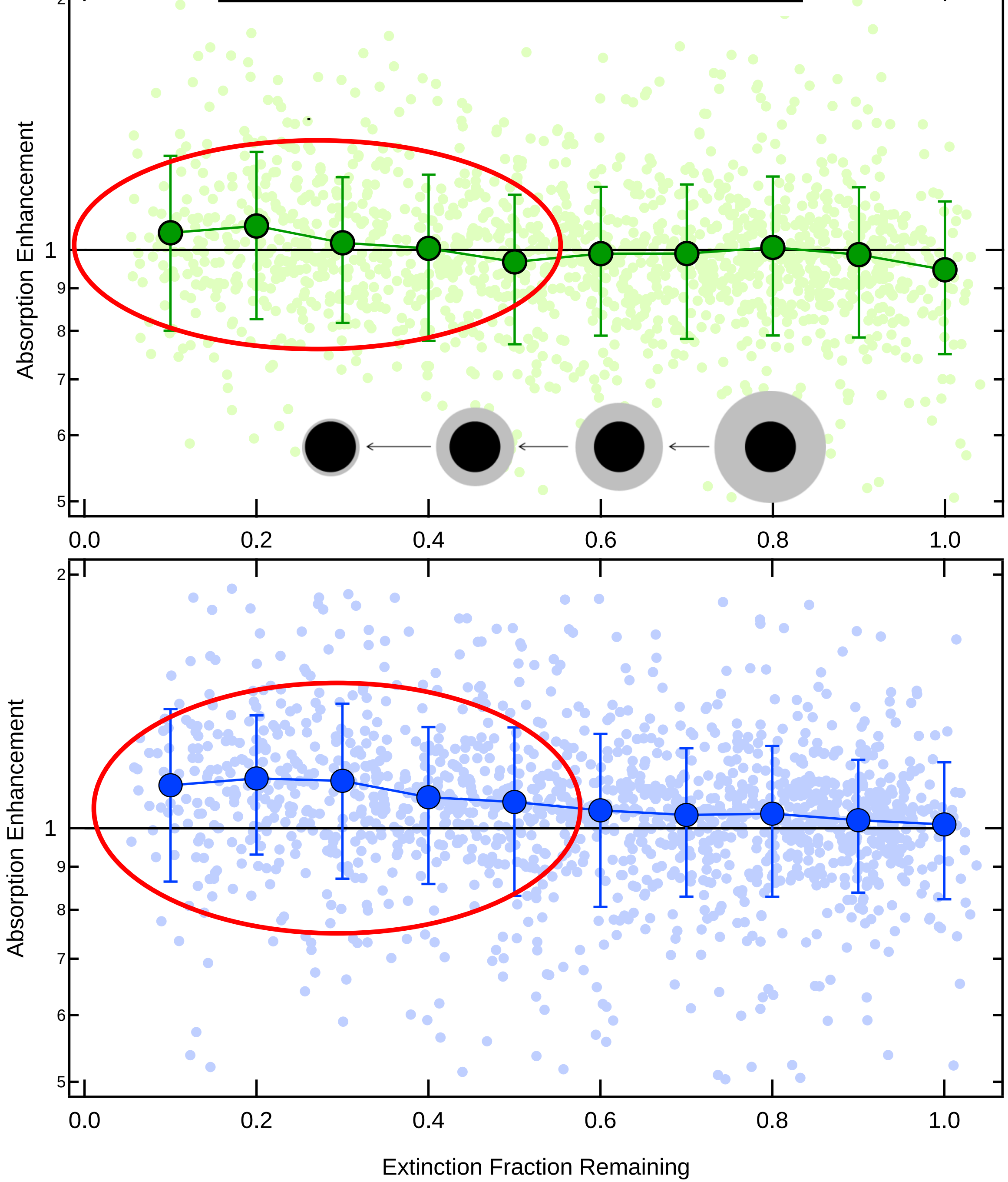



Points = observed
Lines = calculations
$D_{p,core}$ = 40 nm
$D_{p,core}$ = 140 nm
Coating/Core Volume Ratio or SP-AMS Ion Ratio
Ambient Single Scatter Albedo (532 nm)
0
20
40
60
80
0.0
0.2
0.4
0.6
0.8
1.0



SP-AMS sees *only* particles that contain black carbon
SSA shows similar variation with $\frac{I_{coating}}{I_{BC}}$ and $\frac{V_{coating}}{V_{core}}$
which would only be the case if particles are internally mixed.
$D_{p,core}$ = 40 nm
$D_{p,core}$ = 140 nm
Coating/Core Volume Ratio or SP-AMS Ion Ratio
Ambient Single Scatter Albedo (532 nm)
0
20
40
60
80
0.0
0.2
0.4
0.6
0.8
1.0

Brown Carbon Absorption

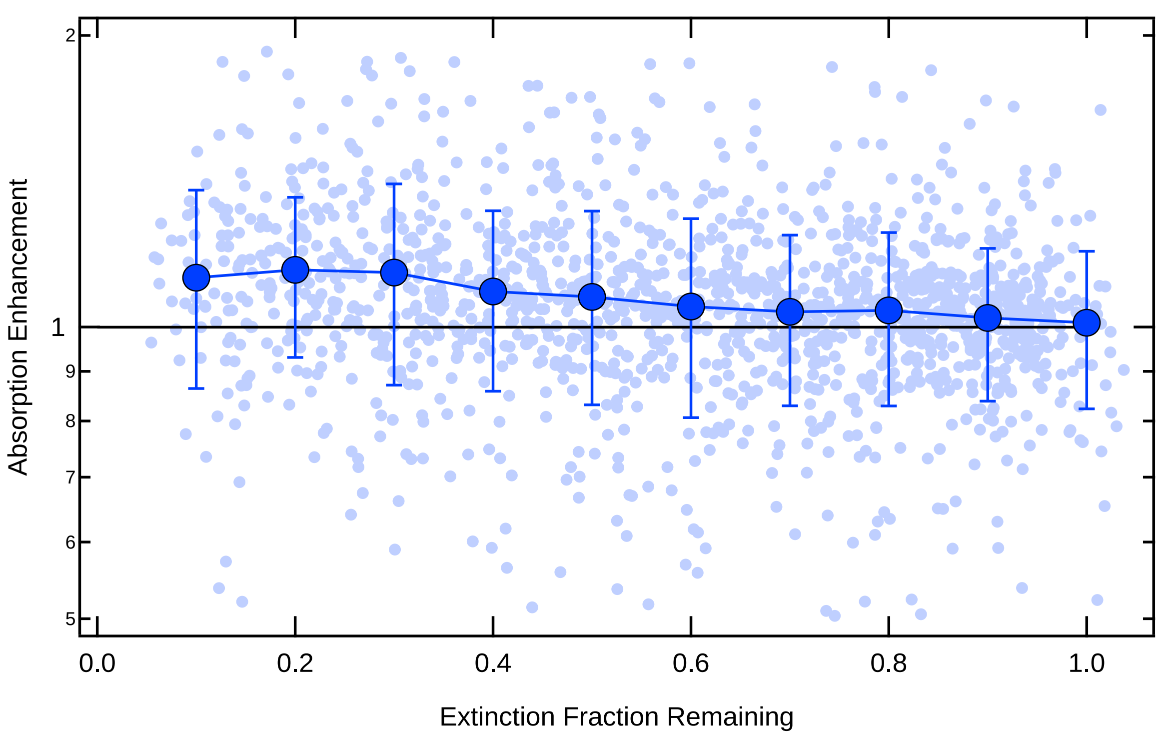


Brown Carbon Absorption

~10% of absorption at 405 nm may be due to “Brown” Carbon

$$MAC\left(\frac{m^2}{g}\right) = \frac{b_{abs}}{m_{OA}} \sim 0.24\,\frac{m^2}{g}$$

Imaginary Refractive Index ~ 0.009



Conclusions

1. Absorption enhancements much smaller than expected
2. Suggests internal mixtures but not with “core-shell” morphology
3. Absorption by “brown” carbon ~ 10% of total at 405 nm

Thanks again to all my great collaborators, the R/V Atlantis Crew and the EPA and NOAA for funding.